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ORIGINAL PAPER

Evaluation of the occurrence of antibiotics at different treatment stages of decentralised and conventional sewage treatment plants

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Abstract

Antibiotics are widely used in human society and are consequently found in sewage treatment plants. However, information regarding the fate of antibiotics in decentralised sewage treatment plants is limited. Therefore, this study monitored and compared four frequently prescribed antibiotics (ampicillin, ciprofloxacin, erythromycin and sulfamethoxazole) in each stage of four conventional treatment plants (three extended aerations and one conventional activated sludge) and two decentralised treatment plants (two Imhoff tanks) in Johor Bahru district between December 2018 and August 2019. The findings revealed that ampicillin, ciprofloxacin, erythromycin and sulfamethoxazole were detected in most wastewater samples, including influent, secondary effluent, final effluent and sewage sludge samples. The antibiotic concentrations in the aqueous and sludge phases ranged from 4.2 to 2690 ng/l and 1.7 to 317.4 ng/g, respectively. Ampicillin and sulfamethoxazole were effectively removed by extended aeration and conventional activated sludge plants (> 75% reduction), erythromycin was effectively removed in IT plants (> 85% reduction) and ciprofloxacin could not be removed effectively by any systems (< 30% reduction). Statistical analysis showed that the approximate relationships between the removal of antibiotics and several wastewater parameters existed. A detailed antibiotic mass flow was conducted in extended aeration and Imhoff tank plants. An estimate of 115.72 g, 202.25 g, 170.55 g and 213.21 g of ampicillin, ciprofloxacin, erythromycin and sulfamethoxazole, respectively, were discharged annually. The mass balance analysis indicated that biodegradation was the major route for the removal of all antibiotics studied. Meanwhile, sorption was only responsible for minor removal of ciprofloxacin, erythromycin and sulfamethoxazole.

Keywords Activated sludge system · Biodegradation · Extended aeration system · Imhoff tank · Mass balance analysis · Sorption

Introduction

The presence of pharmaceutical and personal care products (PPCPs) in the environment has received considerable attention in recent years due to their adverse environmental and human health effects (Behera et al. 2011). As one of the most important subclasses of PPCPs, the global consumption of antibiotics has increased 39% from 2000 to 2015 (Klein et al. 2021), and increased 74.49% from 2010 to 2020 (Aditi et al. 2021). Consumed antibiotics are mainly eliminated from human bodies via faeces and urine, which end up in sewage treatment plant (STP).

Conventional STPs such as activated sludge systems are well established around the world and have served as the main treatment technology for wastewater management. However, these systems were not initially designed to remove antibiotics. Therefore, they have been frequently reported to be inefficient

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in removing the antibiotics. Studies in New Zealand (Kumar et al. 2019), the USA (Gao et al. 2012), Europe (Rodriguez-Mozaz et al. 2020; Drillia et al. 2005) and several Asian countries including China (Hu et al. 2018; Sun et al. 2014), Japan (Matsuo et al. 2011), Korea (Behera et al. 2011) and Singapore (Tran et al. 2016) have observed the incomplete removal of antibiotics by conventional STPs.

Despite the current adoption of the conventional treatment system as the main treatment technology, many countries still employ decentralised treatment systems in urban and semi-rural areas (Nguyen et al. 2018; Capodaglio 2017; Yacob et al. 2017; Istenic et al. 2015) due to economic constraints (Chong et al. 2013) and environmental considerations (Ho and Anda 2006). Therefore, adopting small and decentralised STPs for the treatment of residential and commercial wastewater is the common approach. For example, 25% of the population in the USA is served by decentralised treatment systems and 50% of the STPs in Malaysia are considered small and decentralised (EPA 2000; IWK 2020). This indicates that the treatment of antibiotics is partially reliant on these systems. If antibiotic residuals are not properly treated, their continuous release into the environment could trigger the development of antibiotic resistance bacteria (ARB) and antibiotic resistance genes (ARG) (Qiao et al. 2018; Martinez and Baquero 2017).

To date, while the data on the behaviour and removal of antibiotics in conventional treatment systems (i.e. activated sludge systems) are available, findings regarding decentralised wastewater systems are apparently missing. This study is essential as decentralised systems are still widely employed as the main treatment approach around the world. Furthermore, very limited studies on the subject matter have been conducted in the Southeast Asian region. This paper presents the results of a study conducted to examine and compare the antibiotic removal capability of a decentralised treatment system, that is, an Imhoff tank (IT) and two conventional treatment systems, that is, conventional activated sludge (CAS) and extended aeration (EA) in Johor Bahru district, Malaysia, between December 2018 and August 2019. Four types of antibiotics were investigated based on their globally common usage (WHO 2018), namely ampicillin (AMP), ciprofloxacin (CIP), erythromycin (ERY) and sulfamethoxazole (SMX). The correlation between the removal of these antibiotics and the wastewater parameters was established, and the removal mechanisms were also explored through mass balance analysis.

Materials and methods

Chemicals and consumables

Methanol, acetonitrile, hydrochloride acid and Na_4EDTA (99.8%) were supplied by QRec (Asia). Formic acid and Oasis HLB 3cc Vac Cartridge (60 mg sorbent) were obtained from Fisher Scientific (United Kingdom) and Waters Cooperation (USA), respectively. Antibiotic standards (ampicillin (AMP, 99%; CAS ID: 69-53-4), ciprofloxacin (CIP, 99%; CAS ID: 85721-33-1), erythromycin (ERY, 99%; CAS ID: 114-07-8), sulfamethoxazole (SMX, 99%; CAS ID: 723-46-6)) and the respective internal standard (ampicillin-d5, ciprofloxacin-d8, erythromycin-13c,d3 and sulfamethoxazole-d4) were purchased from Santa Cruz Biotechnology (USA). Stock solutions with 500 mg/L of individual antibiotic were prepared in methanol and stored in the dark at a temperature of − 18 °C before use. The antibiotic stock solutions were diluted for the subsequent work and calibration standards for every sampling event. Milli-Q Advantage A10 System (> 18.2 Ω) was employed for all dilution and sample preparation works. All reagents and chemicals were of analytical grade.

Wastewater sample collection

Samples were collected from six municipal STPs consisting of three EAs (referred to as EA1, EA2 and EA3), two ITs (referred to as IT1 and IT2) and one modular CAS system. The treatment stages for the selected STPs and their respective operational parameters are given in Table 1.

Both wastewater and sludge were collected during three sampling campaigns between October 2018 and August 2019, at the sampling points shown in Figs. 4, 5 and 6. The detailed sampling dates are presented in Tables S1 and S2 (supplementary material). These sampling points were selected to evaluate the occurrence and changes in antibiotic concentrations in different treatment units of the selected STPs. Sludge samples at the EA and IT plants were collected to evaluate the antibiotic concentrations in sludge. Sewage sludge samples of EA plants were collected from the settled sludge of secondary effluent. Sewage sludge samples of IT plants were collected during periodic desludging process. The sludge sample from EA plants used for MLSS measurement was sampled directly from aeration tank. However, the sludge and wastewater samples could not be collected after the aeration stage of the CAS owing to the inaccessibility issue.

One litre of the sample was collected in duplicate from each sampling point using amber glass bottles (prewashed with detergent and distilled water and dried at 400 °C). The collected samples were kept in an ice-box and transported to the laboratory. The samples for wastewater parameters quantification were processed and measured immediately. Meanwhile, the samples for antibiotic detection were immediately filtered through a 0.45-um cellulose nitrate filter (NC, Bioflow) to remove suspended solids, preserved in the freezer and stored at 4 °C before extraction. The extraction was done

Table 1 Treatment stages and operational parameters of selected STPs

STPs	Treatment stages	Designed population equivalent	Flow rate (m^3/d)	Sludge discharge rate (kg/d)	HRT (h)	Dissolved oxygen (mg/L)	Mixed liquor-suspended solid (mg/L)
EA 1	Screening → Aeration → Secondary Clarifier → Final effluent	945	882	1745	29.9	2.46 ± 1.2[1]	793 ± 109
EA 2		1170	805	159	22.9	4.3 ± 2.3[1]	1053 ± 119
EA 3		995	923	183	13.4	3.12 ± 1.2[1]	1120 ± 225
IT 1	Screening → sedimentation → sludge digestion → Final effluent	1140	5.6	0.12	2–4	0.22 ± 0.2[1]	–
IT 2		940	673	6.4	2–4	0.38 ± 0.14[1]	–
CAS	Screening → Equalisation tank → Aeration → Sedimentation	800	1085	215	23.6	2.45 ± 1.3[2]	–

[1]Measured from aeration/treatment tank

[2]Measured from effluent samples

within 24 h after the collection to minimise the hydrolysis and degradation of the antibiotics.

Sewage characterisation

The samples were analysed for wastewater quality parameters according to the standard method (APHA 2017). The analysed parameters include turbidity, chemical oxygen demand (COD) (total and soluble), five-day biochemical oxygen demand (BOD_5), nitrogen (total nitrogen, ammonia, nitrate and nitrite), total phosphorus (TP) and suspended solids (total and volatile, i.e. TSS and VSS, respectively). The temperature and pH were measured in situ using a water quality checker (Horiba, USA).

Sample extraction

All wastewater and sludge samples were extracted, concentrated and analysed according to USEPA Method 1694 (EPA 2007). In brief, 150 ml wastewater samples were acidified with concentrated hydrochloride acid followed by the addition of 500 mg of Na_4EDTA powder. The samples were allowed to rest for 2 h. Accurately measured 20 ng of corresponding internal standards was spiked into all samples. Before extraction, the vacuum cartridge was conditioned with 10 ml methanol and 5 ml distilled water. The samples were extracted through solid-phase extraction (SPE) by allowing 150 ml of wastewater samples to pass through Oasis HLB cartridges (Water, USA) with a flow rate of approximately 1 ml/s. The eluents process was initiated with vacuum pump and finished with gravity to minimise analytes loss. The eluents were concentrated to 1 mL with a nitrogen evaporator (XcelVap; Horizon Technology, USA). Concentrated eluents were filled up to 4 mL using formic acid (0.1% v/v in methanol).

For sludge samples, an ultrasonic extraction method was employed before SPE, as described for the wastewater samples (EPA 2007). Wet sludge samples of 2.5 g were acidified by 15 ml of an acidified phosphate buffer solution and added with corresponding internal standards. Further, the samples were added with 20 ml acetonitrile and placed in an ultrasonic cleaner (UCP-10; Lab Companion, Korea) for 30 min. Upon sonication, the samples were centrifuged at 3000 rpm for 5 min and the supernatants were acidified to pH 2.0 and transferred to a round-bottom flask. Similar procedures were repeated thrice for the centrifuged sludge to ensure the complete extraction of the desired analytes. It is important to note that the phosphate buffer solution was replaced with acetonitrile in the final extraction. The supernatant was concentrated using a rotary evaporator (Rotavapor R-210; Buchi, Switzerland) to the final volumes of 30 ml and 200 ml of ultrapure water and 500 mg Na_4EDTA was immediately added. The samples were then processed using the SPE procedures, as described previously.

Instrumental analysis

The concentrations of antibiotics and internal standards were determined according to USEPA Method 1694 (Isotope Dilution Method) (EPA 2007) with an Ultra-High-Performance Liquid Chromatography system, 1290 Infinity (Agilent, USA), coupled with an Electrospray Triple Quadrupole Mass Spectrometer (6410; Agilent, USA). Zorbax Eclipse Plus C18 Rapid Resolution HD column (2.1 × 50 mm, 1.8 µm; Agilent, USA) was used to separate the desired compounds at a flow rate of 0.5 ml/min and temperature of 30 °C. Two mobile phases were employed for the compound separation, namely, formic acid with a concentration of 0.1% in water (v/v) (mobile phase A) and 0.1% of formic acid in acetonitrile (v/v) (mobile phase B). The gradient elution method was utilised wherein the mobile phase A was started at 95% and linearly decreased to 70% in the first minute, followed by a further decrease to 55% in the second minute. It was finally reduced to 0% in the next 0.5 min and held until the end of each run.

The MS system was operated in the positive ionisation mode with particular operating conditions: a gas temperature of 300 °C, a capillary voltage of 4 kV, a gas flow rate at 11 L/min and gas pressure at 45 psi. The specific details of multiple reaction monitoring for the studied antibiotics are outlined in Table S3. The injected sample volume was 1.0 µL.

Quantification and quality control

In this work, a nine-point calibration curve (0.01–1000 ng/mL) was generated with strong linearity ($R^2 > 0.999$) for all targeted antibiotics with corresponding internal standards. During the analysis, one blank and one independent check standard were run before the injection of the next sample to ensure there was no background contamination and for the system performance check. Data acquisition and analyses were carried out by the Agilent MassHunter Workstation Software.

The method detection limit (MDL) was estimated from the analyses of ten blank samples with target antibiotics spiked in ultrapure water. The MDL for the analytes was calculated by the formula $3 \times \mathrm{SD/m}$, where SD is the standard deviation of the lowest signal/noise ratio of the analyte and m is the slope of the calibration curve. Further, the MQL was calculated as $10 \times \mathrm{SD/m}$ (Tran et al. 2013). The MDL and MQL for AMP, CIP, ERY and SMX were 20 and 65, 1.4 and 4.5, 0.8 and 2.5, and 1.3 and 4.0 ng/L, respectively. Extraction recoveries were done for all antibiotics to inspect the performance of this method through the isotope dilution method (EPA 2007). The isotope label compound recoveries for all antibiotics were within the range of acceptance. The results and details for antibiotic recoveries are listed in Table S4.

Data analysis

Statistical analysis

Non-parametric Kruskal–Wallis (KW), Mann–Whitney (MW), ANOVA and Pearson correlation (P) tests were performed using OriginLab 2017. The non-parametric Kruskal–Wallis test was used to determine the significant differences between three groups of data, Mann–Whitney test was used to determine the significant differences between two groups of data and ANOVA was used to inspect the solidity of the relationship. Spearman's correlation was used to determine the relationship between the removal of wastewater parameters and antibiotics. Spearman's correlation of more than 0.7 was considered as significantly correlated and vice versa (RGC-IBG n.d.). The p-values from Kruskal–Wallis, Mann–Whitney and ANOVA tests were treated as insignificant if they were greater than 0.05.

Preliminary mass balance analysis

Mass balance analysis was used to determine the fate of antibiotics in each treatment unit during the treatment process. This allowed us to understand the process that contributes the most towards the removal of antibiotics. The mass balance analysis was conducted through the mass flow approach, in which the concentration of compounds was converted to mass flow (g/day). The volatilisation of antibiotics was considered as insignificant during the treatment process (Joss et al. 2006); hence, only biodegradation and sorption were analysed. The procedures of mass balance analysis followed the work of Carballa et al. (2007) and Gao et al. (2012).

Initially, the measured concentrations of each antibiotic in the aqueous and sludge phases were converted to mass flow (MF) using Eqs. (1) and (2), respectively.

$$\mathrm{MF_{aq}} = \mathrm{C_{aq}Q_{aq}} \tag{1}$$

$$\mathrm{MF_{s}} = \mathrm{C_{s}Q_{s}} \tag{2}$$

In the above equations, $\mathrm{MF_{aq}}$ and $\mathrm{MF_{s}}$ (ng/d) are the calculated mass flows of the antibiotic in the aqueous and sludge phases, respectively; $\mathrm{C_{aq}}$ (ng/L) and $\mathrm{C_{s}}$ (ng/g) are the soluble average concentrations of the antibiotic in the aqueous and sludge phases, respectively; and $\mathrm{Q_{aq}}$ ($\mathrm{m^3/d}$) and $\mathrm{Q_{s}}$ (kg/d) are the average flow rate of wastewater and discharge rate of sludge in the treatment plant, respectively.

From the above, the total mass flow ($\mathrm{MF_t}$) (kg/d) of individual antibiotics is calculated using Eq. (3). This allowed us to estimate the mass of antibiotics treated and discharged from the STPs.

$$\mathrm{MF_t} = \mathrm{MF_{aq}} + \mathrm{MF_{s}} \tag{3}$$

Equation (4) was used to determine the fraction of antibiotics biodegraded during the treatment process.

$$\mathrm{MF_{bio}} = \mathrm{MF_{aq,inf}} - \left(\mathrm{MF_{aq,eff}} + \mathrm{MF_{s}}\right) \tag{4}$$

In the above equations, $\mathrm{MF_{aq,inf}}$ (kg/d) and $\mathrm{M_{aq,eff}}$ (kg/d) are the mass flows of the antibiotic in the aqueous phase of influent and effluent of the treatment system and $\mathrm{M_{bio}}$ (kg/d) is the mass of the antibiotic lost due to biodegradation.

Equations (5) and (6) were used to express the fraction of antibiotics that were biodegraded ($\mathrm{F_{bio}}$) and sorbed ($\mathrm{F_{s}}$) in percentage form:

$$\mathrm{F_{bio}} = \frac{\mathrm{MF_{bio}}}{\mathrm{MF_{Total}}} \times 100 \tag{5}$$

$$\mathrm{F_{s}} = \frac{\mathrm{MF_{s}}}{\mathrm{MF_{Total}}} \times 100 \tag{6}$$

In the above equations, M_{total} is the total mass of the antibiotic lost during the treatment process, obtained from Eq. (3).

Finally, to evaluate the capability of sorption into the sludge of an individual antibiotic, a solid–liquid distribution coefficient (K_d) was used as an estimation using Eq. (7) (Xue et al. 2010), as follows:

$$K_d = \frac{C_s \,.MLSS}{C_{aq}} \quad (7)$$

In the above equation, MLSS is the concentration of the mixed liquor-suspended solid in the aeration chamber (g/L) and K_d (L/g-MLSS) is the solid–liquid distribution coefficient of the individual antibiotic.

Results and discussion

Occurrence of antibiotics

Wastewater

The occurrences and detected concentrations of antibiotics in each STP are shown in Table 2. All antibiotics were detected in influent, secondary effluent and final effluent samples. The detection frequencies of antibiotics in influent samples are in the following order: CIP: 66.67% > AMP: 55.56% = SMX: 55.56% > ERY: 31.11%. The antibiotic concentrations detected in all STPs ranged from 149.4 to 2,690 ng/l in influent samples, 101.2 ng/l to 858.9 ng/l in secondary effluent samples and 4.2 ng/l to 1,260 ng/l in final effluent samples. In general, as the wastewater was treated, the antibiotic concentration decreased, but could still be detected in the final effluent samples.

Among the four antibiotics, CIP had the highest detection frequency in all influent samples. The high detection frequency of CIP could be related to the widespread use

Table 2 Average concentrations and removals of the antibiotics in the STPs

Antibiotics	EA 1	EA 2	EA 3	IT 1	IT 2	CAS
Influent (ng/L) ($n=3$)						
AMP	2,690.4*	373.1*	152.6 ± 62.4	161.1 ± 207.5	568.3 ± 124.3	182.9*
CIP	450.2 ± 469.8	149.4 ± 106.2	165.1 ± 140.3	155.6 ± 186.6	164.8*	167.6*
ERY	337.1 ± 135.9	587.2*	< MQL	329.5 ± 275.7	205.6*	< MQL
SMX	1,782.3 ± 1,642.6	941.3 ± 945.8	962.3 ± 862.2	916 ± 1241	174.8*	1764.7*
Secondary effluent (ng/L) ($n=3$)						
AMP	188.6*	153.9*	101.2 ± 18.8	NA	NA	NA
CIP	120.2 ± 103.2	116.7 ± 130	152.7 ± 89.6	NA	NA	NA
ERY	495.4*	305.3*	< MQL	NA	NA	NA
SMX	858.9 ± 433.8	294.5*	118.2 ± 38.1	NA	NA	NA
Final effluent (ng/L) ($n=3$)						
AMP	71.4*	27.8*	27.3 ± 19.5	133.3 ± 124.8	305.9 ± 58.3	4.2*
CIP	246.7 ± 154.1	97.3 ± 90	127.5 ± 73.2	111.5 ± 141.9	208.2*	178.9*
ERY	299.9 ± 101.8	211.3*	< MQL	193.7*	44.9*	< MQL
SMX	1264.8*	288.5*	190.1 ± 32.1	569.1 ± 792.8	91.9*	391.3 ± 257.3
Sewage sludge (ng/g)						
AMP	ND	ND	ND	ND	ND	NA
CIP	317.4	140.9	82.1	24.1	51.9	NA
ERY	22.5	17.5	ND	6.5	8.8	NA
SMX	29.7	40.2	26.9	1.7	3.9	NA
Overall removal (%)						
AMP	97.3*	92.6*	85.7 ± 8.4	40.8 ± 3.5	58.9 ± 21.2	97.7*
CIP	24.1 ± 17.8	40.9 ± 7.6	8.6 ± 23.5	39.6 ± 13.3	− 26.3*	− 6.8*
ERY	9.8 ± 4.4	64*	NA	89.9 ± 10.1	78.1*	NA
SMX	78.5 ± 17.6	94.9 ± 5.1	64.5 ± 24.8	57.5 ± 20.4	47.4*	77.8*

MQL, method quantification limit; NA, data not available, ND, not detected

*Data without ± SD represented less than two samples detected

of this antibiotic for antibacterial purposes by the Malaysian health facilities. According to the Ministry of Health of Malaysia (MOH), CIP is one of the top 50 drugs in the Malaysian public and private sectors with a sale of more than RM 18.9 million (MSOM 2014). Conversely, the lower detection frequency of AMP, ERY and SMX in all influent samples could be due to the lesser consumption of these drugs as these antibiotics are not heavily used in Malaysia (MSOM 2014). It was also observed that ERY was not detected in all samples from the CAS plant, indicating that it may not be consumed by the community within the CAS catchment.

The influent concentration of SMX (1090 ± 1000 ng/l) was the highest among all antibiotics, followed by AMP (491.1 ± 254.3 ng/l), ERY (350.8 ± 203.8 ng/l) and CIP (231.1 ± 723.6 ng/l). The influent concentrations of SMX and ERY in this work were higher compared to studies from European countries (Papageorgiou et al. 2016; Göbel et al. 2005), but consistent with studies from Southeast Asian countries (Tran et al. 2016). The difference in the detected antibiotic concentrations in influents from different studies may be due to the difference in drug prescription practices between the regions (Akiba et al. 2015).

In the secondary effluents, SMX appeared as the highest concentration antibiotic (449.8 ± 392.1 ng/L), followed by ERY (400.4 ± 95.1 ng/L), AMP (136.3 ± 38.3 ng/L) and CIP (128.5 ± 82.7 ng/L). In the final effluent samples, SMX remained the highest concentration antibiotic, having a detected concentration of 398.1 ± 443.3 ng/L, followed by ERY (209.9 ± 103.9 ng/L), CIP (158.1 ± 105.3 ng/L) and AMP (115.1 ± 120.2 ng/L). These findings showed that the concentrations of selected antibiotics remained above 100 ng/L throughout the treatment line.

Sewage sludge

The antibiotics and their concentrations detected in the sewage sludge are shown in Table 2. In general, most antibiotics were detected in all samples, except for AMP. Of these, CIP had the highest concentration (24.1 ng/g to 317.4 ng/g) compared to ERY (6.5 ng/g to 22.5 ng/g) and SMX (1.7 ng/g to 40.2 ng/g). The antibiotic concentrations in the sludge from EA plants were relatively high compared to IT plants. The concentrations of ERY and SMX in the sewage sludge from EA plants were similar to previous studies (Hu et al. 2018; Gao et al. 2012) conducted in activated sludge systems. However, the CIP concentration detected in this study was higher than in previous studies (Hu et al. 2018; Li et al. 2013). No comparison could be made for IT plants as no study has been conducted on IT plants elsewhere.

Removal efficiencies of antibiotics in STPs

The overall removal efficiencies of all antibiotics in different treatment systems are shown in Fig. 1. Of these, CIP (− 26.4% to 59.7%) had a significantly lower removal efficiency as compared to AMP (29.3% to 97.7%), ERY (5.4% to 99.9%) and SMX (39.7% to 99.9%) (MW, $p < 0.05$). The findings from this study allowed us to know that CIP was more resistant to treatment regardless of the treatment system employed when compared to AMP, ERY and SMX. The negative removal could be the result of the transformation of the conjugated compound to parent compound by microorganisms during the treatment process (Göbel et al. 2005). The negative removal of CIP was reported in a previous study with removal as low as − 88.6% (Blair et al. 2015).

The comparison of antibiotic removals between this study and previous studies is shown in Fig. 2. In general, the overall removal of AMP and CIP in this study is considerably different with previous studies. However, the removals of ERY and SMX are similar with previous studies. Between 56.8 and 95% of AMP removal was reported in previous studies (Papageorgiou et al. 2016; Mutiyar and Mittal 2014; Li and Zhang 2010), but only 29.3% to 97.7% removal was recorded in this study. As for CIP, lower removal was achieved by the treatment plants in this work. In previous studies, − 4.5% to 96.1% removal of CIP were recorded (Min et al. 2018; Li et al. 2013; Li and Zhang 2010), but only − 26.4% to 59.7% of removal was recorded in this work.

The removal efficiencies of ERY in this work are slightly higher compared to previous studies. About 5.4% to 99.9% of ERY was removed in this work, while 0 to 90% of ERY removal was reported by previous studies (Hu et al. 2018; Li et al. 2013; Li and Zhang 2010; Göbel et al. 2005). On the other hand, SMX have the similar removal between

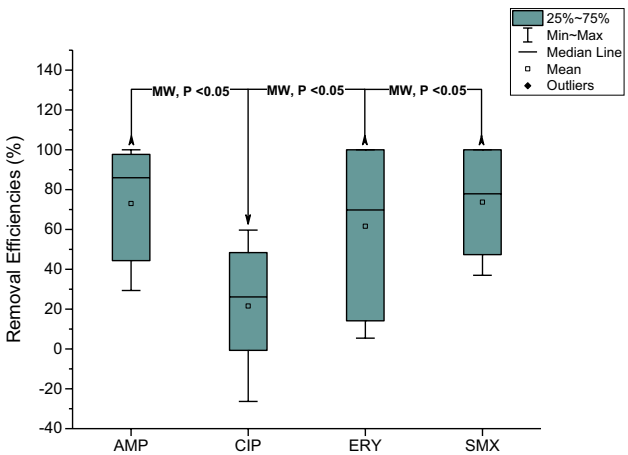

Fig. 1 Overall removal efficiencies of target antibiotics in all STPs

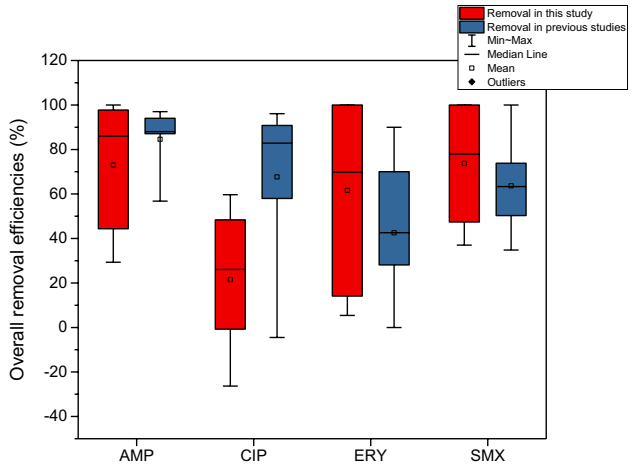

Fig. 2 Comparison of overall antibiotic removal efficiencies between this study and previous studies

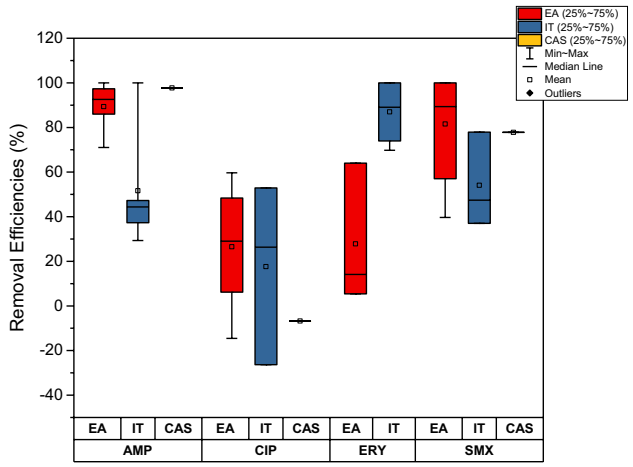

Fig. 3 Removal efficiencies of antibiotics in different system

this study (39.7% to 99.9%) and previous studies (34.8% to 97.3%) (Min et al. 2018; Göbel et al. 2005).

The removal of antibiotics according to the treatment system used is shown in Fig. 3. In general, EA plants achieved slightly better antibiotic removal efficiencies as compared to IT and CAS plants. CIP and SMX were better removed in EA plants; on the other hand, only ERY was better removed in IT plants and only AMP was better removed in CAS plants. However, the removal of only ERY was significant between EA and IT plants (CAS was excluded as it was undetected in the sample) (MW, $p < 0.05$), and AMP, CIP and SMX were insignificantly different between all studied treatment systems (KW, $p > 0.05$). This indicates that the removal of AMP, CIP and SMX was similar in the studied plants. The average removals of the targeted antibiotics were as follows (EA vs IT vs CAS): AMP (89.4 ± 11.6% vs 51.6 ± 27.9% vs 97.4%), CIP (26.5 ± 26.7% vs 17.6 ± 40.3% vs − 6.8%), ERY (27.9 ± 31.6% vs 86.9% ± 15.4% vs undetected) and SMX (81.6 ± 23.9% vs 54.1 ± 21.3% vs 77.8%). The negative removal of CIP was observed in all treatment systems. The results indicated that different types of systems seemed to have different capabilities in removing different types of antibiotics.

AMP was the only antibiotic that could effectively be removed by all the studied systems. As one of the β-lactam class antibiotics, the effective removal of AMP has been frequently reported. Matsuo et al. (2011) and Tran et al. (2016) concluded that AMP was effectively removed in conventional STPs, with removal efficiencies up to 90% and 87%, respectively. The high removal of AMP could be due to the easy hydrolytic cleavage of the β-lactam ring within the AMP to CO_2 and water by β-lactamase, which is a common and widespread enzyme in the wastewater (Yagiela et al. 2010).

The resistance to treatment characteristic of CIP was observed in this work. The removal of CIP was below 60% regardless of the system employed. The removal of CIP recorded in this study contradicts most of the previous findings. Although one previous study showed the negative removal of CIP (Blair et al. 2015), the majority of studies showed the effective removal of CIP with an average removal of more than 70% (Hu et al. 2018; Min et al. 2018; Tran et al. 2016; Dorival-García et al. 2013; Li and Zhang 2010). Of these, Li and Zhang (2010) and Dorival-García et al. (2013) reported consistent results regarding the effective removal of more than 90% of CIP in the activated sludge system. Similarly, Tran et al. (2016) reported the removal of CIP with an efficiency of 86% in a full-scale CAS system.

The differences between the results of this work and previous studies could be due to two reasons. First, the MLSS concentrations of EA plants in this work are significantly lower compared to the activated sludge plants in other works. In previous studies, the reported MLSS was approximately 2300 to 2500 mg/L (Kang et al. 2018; Tran et al. 2016). However, the MLSS of EA plants in this work was only 793 to 1120 mg/L (Table 1). Second, the poorer removal of CIP could be due to the absence of a mixing mechanism in IT plants. CIP has often been reported to be removed through sorption onto biomass due to the cation characteristic of CIP under neutral pH conditions (Tran et al. 2016; Dorival-García et al. 2013; Li and Zhang 2010). While the biomass concentration and mixing have been reported to have a positive effect on the sorption of compounds (Kim et al. 2005), the lower MLSS concentration in the EA plants and absence of mixing in the IT plants might have reduced the sorption of CIP onto the biomass, and hence the removal.

ERY was only detected in samples from EA and IT plants. The removal of ERY was more effective IT plants (69.8% to 78.1%) as compared to EA plants (5.4% to

64.1%). Furthermore, the performance was also more stable in IT plants, judging from its low variation. The effective removal of ERY in IT plants might be due to the anaerobic conditions that prevailed in the system. According to Xue et al. (2010), the removal of ERY was better under anaerobic conditions and less effective under aerobic conditions. Furthermore, previous findings on the removal of ERY in activated sludge systems were inconsistent. Some studies showed that ERY was ineffectively removed (< 60%) (Tran et al. 2016; Li and Zhang 2010; Göbel et al. 2005), while other reported the opposite (> 85%) (Suárez et al. 2010). The reason for this inconsistency is currently unknown and requires further examination.

The removal of SMX was more effective in EA and CAS plants (> 75%) as compared to IT plants (< 55%). The differences in pollutant removal might be due to the different redox conditions. SMX was reported to be effectively removed in aerobic conditions compared to anaerobic conditions (Kang et al. 2018; Zhao et al. 2018). Kang et al. (2018) reported that SMX was not removed under anaerobic conditions and was only removed after the reactor changed to aerobic conditions. Furthermore, as SMX was mainly removed through biodegradation (Min et al. 2018; Kang et al. 2018), aerobic conditions, which are significantly related to nitrification, were reported to enhance the removal of SMX (Kang et al. 2018; Alvarino et al. 2014).

Mass flow of antibiotics

Figures 4, 5 and 6 show the mass flow of antibiotics for three EA plants (EA1, EA2 and EA3), two IT plants (IT1 and IT2) and the CAS plant, respectively. The mass flow was the mass fraction of antibiotics that left each treatment unit through its stream. The data for antibiotics in the sewage sludge of the CAS plant were unavailable as the sample was inaccessible. In general, the mass fraction of antibiotics decreased throughout the process. The mass fraction of antibiotics in the secondary effluent and final effluent samples of all plants ranged from 7 to 127% and 2.2 to 126.3% of the influent mass, respectively, indicating that a high mass fraction of antibiotics was discharged into the receiving river after the treatment process.

In the EA process, the mass fraction of antibiotics found in the stream after the aeration tank varied; large inconsistencies were observed between antibiotics and even between the plants (Fig. 4). The lowest mass fraction found in the secondary effluents was 7%, which was for AMP in EA1, and the highest mass fraction found in secondary effluent was ERY in EA1, which was 146.9%. For AMP, 41.2% to 69.1% of the influent mass was found in the secondary effluents of EA2 and EA3, but a lower fraction (7%) of the AMP mass fraction was found in the secondary effluent of EA1. A similar trend was observed for CIP, in which 78.1% to 92.5% of CIP was found after aeration tank in EA2 and EA3, but a lower fraction (27%) was found in the secondary effluent of

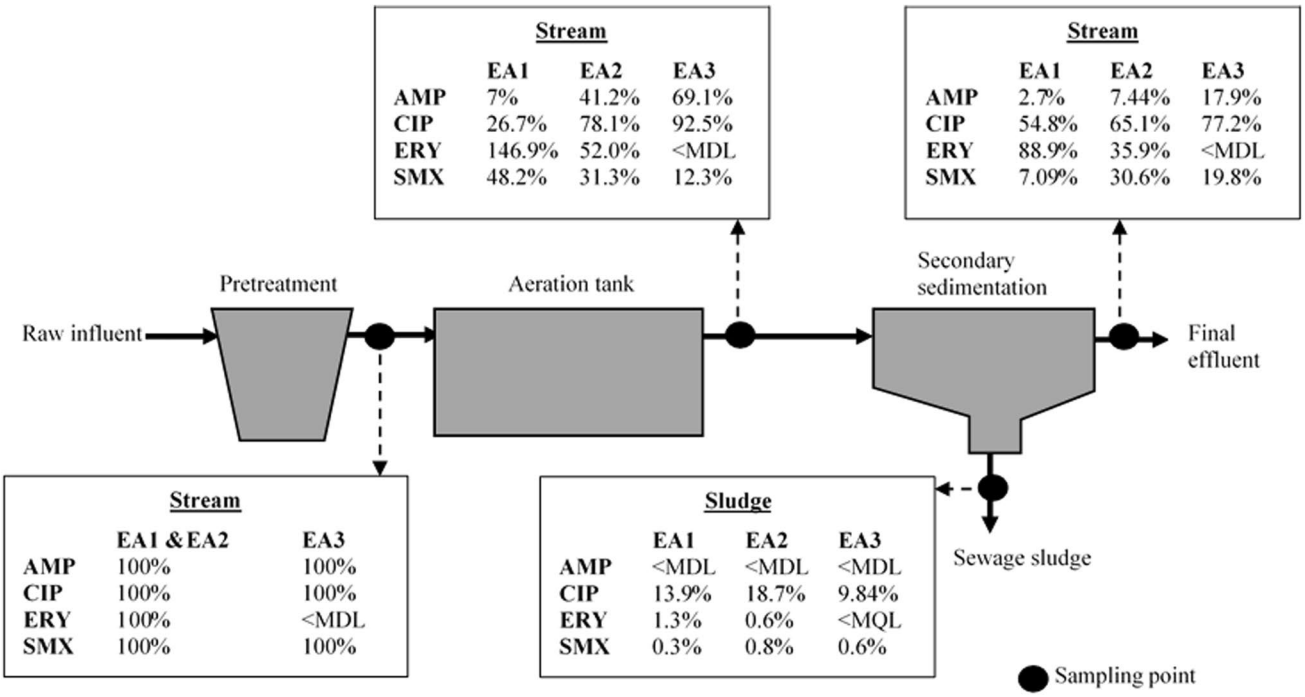

Fig. 4 Mass flow of antibiotics in EA plants. *MDL* method detection limit

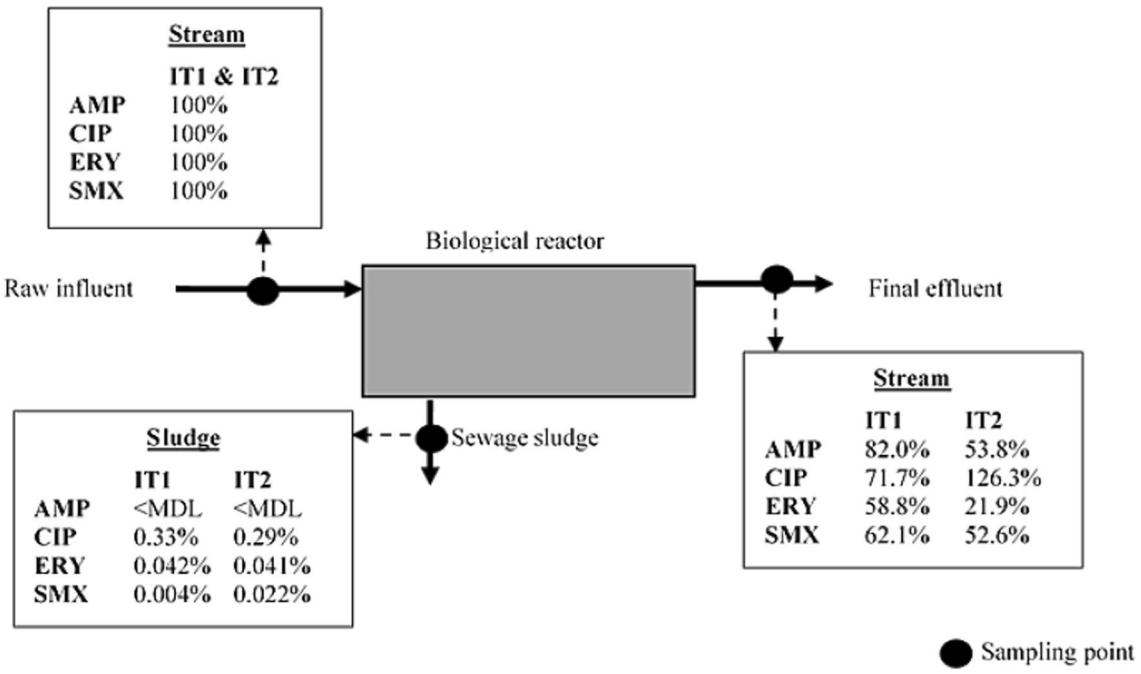

Fig. 5 Mass flow of antibiotics in IT plants. *MDL* method detection limit

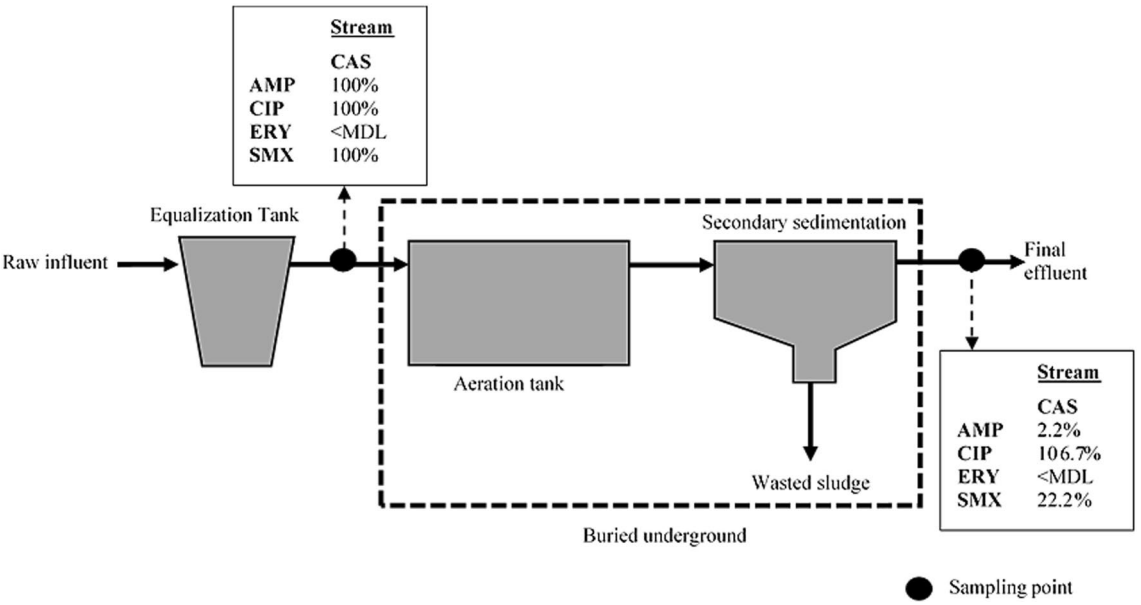

Fig. 6 Mass flow of antibiotics in CAS plant. *MDL* method detection limit

EA1. An increase in the mass fraction after the aeration tank process was observed for ERY; 52% to 147% of ERY was detected in the secondary effluents of EA plants. The mass fraction of SMX found in the secondary effluents was higher in EA1 (48%) and EA2 (31%) as compared to EA3 (12%).

The secondary clarifier in EA plants further decreased the mass fraction of antibiotics in the secondary effluents. Less than 30% of the original AMP (2.7% to 17.9%) and SMX (7.09% to 30%) were found in final effluents of all EA plants; however, a slight increase of the SMX mass fraction was observed in EA3 after the secondary clarifier was utilised. The mass fraction of CIP also increased after the secondary clarifier was implemented in EA1. A

consistent ERY mass fraction was found in the final effluents of EA1 and EA2 (35% to 88%).

In the IT systems, the mass fractions of AMP, SMX and ERY decreased after passing through the treatment chamber, with mass fractions left in final effluents ranging from 2.2 to 82% (Fig. 5). For CIP, the mass fractions in both IT plants increased after treatment (106.7% and 126.3%). The lowest mass fraction found after treatment was for AMP in IT2, with only 2.2% of the influent mass detected.

The increase in the mass fraction after passing through the treatment unit was commonly reported (Gao et al. 2012). However, the reason for the negative removal of antibiotics in STPs is currently unclear. Several hypotheses have been postulated, such as (1) microorganisms break down the faecal particles in the wastewater and released the PPCPs that were initially enclosed within these particles (Göbel et al. 2005); (2) undetected metabolites are transformed to their parent compounds by microorganisms (Blair et al. 2015) and (3) as the antibiotic concentration decreases in the wastewater, the desorption of the antibiotic from the sludge into water phases takes place when the sorption equilibrium shifts to the aqueous phase; this is very much related to antibiotics that possess strong sorption potential (Gao et al. 2012).

The mass fractions of antibiotics that were detected in the sewage sludge from EA plants were significantly higher as compared to the sludge from IT plants. In EA plants, 0.6% to 18.7% of antibiotics were sorbed into the sludge, while only 0.004% to 0.3% of antibiotics were found in the sludge from IT plants. Among all antibiotics, CIP had the highest mass fraction that sorbed into sewage sludge in both systems.

The findings from this section revealed that up to 117.4 g, 273.1 g, 170.6 g and 734.6 g of AMP, CIP, ERY and SMX, respectively, are discharged into the receiving river annually from all the STPs studied in this work. Although this amount could be insignificant, the situation may be amplified in larger-scale plants with greater flow rates.

Mass balance analysis of antibiotics

The removal pathways of antibiotics in EA and IT plants were estimated using the principle of mass balance ($MF_{bio} = MF_{aq,inf} - [MF_{aq,eff} + MF_{sorp}]$. The mass fractions of the antibiotic that was biodegraded were estimated using Eqs. (4) to (6). The mass fractions that were removed through sorption were estimated through the direct measurement of antibiotics in the sewage sludge. The mass fractions of antibiotics that were removed through biodegradation and sorption are illustrated in Fig. 7.

AMP was fully removed through biodegradation; sorption was absent throughout the removal process in EA and IT plants. The fractions of biodegraded AMP in EA plants were large, ranging from 82.1 to 91.3%. This finding supports the

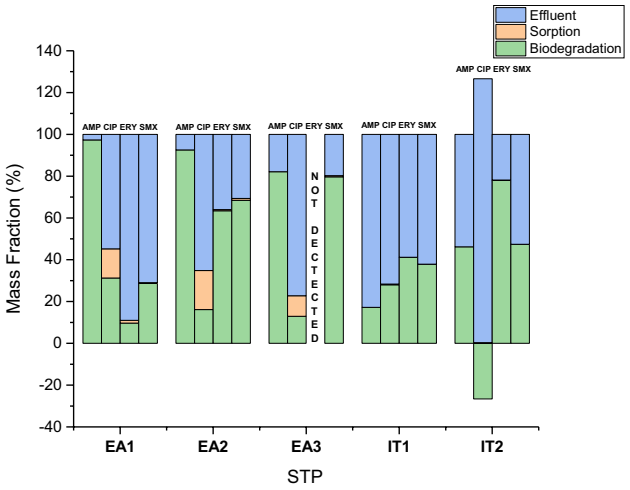

Fig. 7 Relative contribution of biodegradation and sorption in removal process

earlier discussion that AMP is highly biodegradable, mainly because of the easily hydrolysed characteristic of this antibiotic (Yagiela et al., 2010). Similar findings were reported by Gao et al. (2012), in which 95% of AMP was removed in activated sludge systems through biodegradation, and less than 0.1% of AMP was found in the wasted sludge.

However, the mass fractions of AMP that were biodegraded in the anaerobic IT plants were much lower as compared to EA plants, ranging from 17.2 to 46.2%. The lower biodegradation of AMP in IT plants could be due to the differences between the redox conditions and microorganism communities in these systems. Previous studies reported that the anaerobic process could not effectively remove AMP during the treatment of wastewater (Huang et al. 2018; Zhou et al. 2006). Instead, effective removal of AMP was reported in previous work under aerobic conditions (Gao et al. 2012). Furthermore, the nitrification process that only occurred under aerobic conditions showed positive effects in the removal of AMP (Muñoz et al. 2020; Labinghisa and Rollon 2014). Besides, certain microorganisms such as *Thiothrix spp.*, *Acinetobacter spp.* and *Flavobacterium spp.* that are related to the degradation of AMP only live under aerobic conditions (Xia et al. 2012; Shen et al. 2010). Hence, it is well understood that the biodegradation and removal of AMP in IT plants were not as effective as in EA plants.

In EA plants, the fractions of CIP that were removed through biodegradation and sorption ranged from 12.9 to 31.2% and 9.8 to 18.6%, respectively. This indicated that both mechanisms were important during the removal process in activated sludge systems. The biodegradability of CIP is well supported by findings reported in previous studies (Min et al. 2018; Wang et al. 2017; Dorival-García et al. 2013; Li and Zhang 2010). The high sorption potential of CIP in activated sludge systems has also been observed in previous

research (Min et al. 2018; Dorival-García et al. 2013); it has been associated with CIP's physicochemical characteristics as a highly polarised and highly water-soluble antibiotic, which can act as a cation (Tran et al. 2016; Dorival-García et al. 2013). Hence, hydrophobic and electrostatic interactions were significant during the removal process in activated sludge systems.

The removal of CIP in IT plants was mainly due to biodegradation (− 26.6% to 28%), with sorption being insignificant (0.29% to 0.33%). The negative mass fraction of CIP was observed only in IT2 as a higher concentration was detected in effluents than in influents. A similar observation was reported by Liu et al. (2017) and has been discussed earlier. Except for the negative value, the biodegradation mass fractions of CIP in IT plants are almost similar to those in EA plants. This indicated that the differences between the systems do not impose significant effects on the biodegradation of CIP. The differences in the sorption behaviour of CIP in IT plants could be caused by the absence of a mixing mechanism in IT plants. The absence of mixing could affect bulk mass transfer and, thus, induce a significant barrier for the sorption of CIP (Wang et al. 2017).

In the case of ERY, biodegradation acted as the primary removal route and sorption was considered insignificant in EA plants. Approximately 9.7% to 63.4% of ERY was biodegraded, and 0.6% to 1.3% of ERY was sorbed into the sewage sludge in EA plants. The high biodegradability of ERY was also reported by Suárez et al. (2010). Suárez et al. (2010) reported that 85% of ERY was removed through biodegradation at a high biodegradation rate in activated sludge systems. The weak sorption behaviour of ERY in activated sludge systems was reported by Hu et al. (2018); the concentration of ERY detected in the sewage sludge was consistent with the results of the present work and was insignificant.

Similarly, biodegradation acted as significant removal route for ERY and sorption was insignificant in IT plants. Approximately 41.2% to 78.1% of ERY was biodegraded and 0.04% was sorbed into the sewage sludge in IT plants. However, the mass fractions of ERY that were biodegraded in the anaerobic IT plants were higher and more consistent as compared to EA plants. The difference between the redox conditions in EA and IT plants might explain this finding as previous studies revealed that ERY was significantly removed under anaerobic conditions and less significantly removed under aerobic conditions (Xue et al. 2010). Furthermore, the denitrification that occurred at a low dissolved oxygen level had a positive effect on the biodegradation of ERY (Polesel et al. 2017). Regarding the low sorption potential of ERY in all plants, despite the physiochemical characteristics of ERY that were expected to favour high sorption, both the present study and previous studies observed the low sorption potential of ERY (Göbel et al. 2005). Hence, further investigation is required to clarify this observation.

The primary removal pathway of SMX was biodegradation (28.7% to 79.7%), with sorption (0.3% to 0.8%) being insignificant in EA plants. The findings of the present study are consistent with previous studies (Min et al. 2018; Alvarino et al. 2014; Suárez et al. 2012, 2010; Joss et al. 2006). For example, Gao et al. (2012) reported that more than 80% of SMX was removed by the CAS system and only 0.1% of SMX was found in the sludge, indicating that a majority of the SMX was removed through biodegradation.

Lower mass fractions of SMX were biodegraded in IT plants as compared to EA plants, ranging from 37.8 to 47.3%. Insignificant fractions of SMX were sorbed into the sewage sludge (0.04% to 0.2%). The removal of SMX was reported to be more effective under aerobic conditions as compared to anaerobic conditions (Kang et al. 2018; Zhao et al. 2018). Studies also revealed that SMX was significantly correlated with nitrification activities; it could be used as a co-subtract during the cometabolism of ammonia (Müller et al. 2013). The low sorption potential of SMX could be due to the characteristics of SMX, as it is neutral and highly water-soluble. Thus, hydrophobic and electrostatic interactions are unlikely to happen (Tran et al. 2016).

Table S5 shows the sorption coefficients of CIP, ERY and SMX that were calculated using Eq. (7). The K_d values validated the sorption potential of CIP being the highest among the targeted antibiotics, followed by SMX and ERY. The K_d values in this work were generally higher as compared to those from previous studies.

A stronger sorption potential of CIP was demonstrated in all systems as compared to ERY and SMX, indicating that the physiochemical characteristics of antibiotics significantly affected their K_d values. Besides, the K_d values of all antibiotics were also significantly higher in EA plants as compared to IT plants, showing that the operation of the treatment systems could also impose significant effects on the sorption coefficient of antibiotics. Furthermore, the K_d values of antibiotics differed significantly within the same treatment system types (e.g. CIP in EA1, EA2 and EA3). Hence, it could be inferred that the sorption of antibiotics onto the biomass in the treatment systems was affected by both internal (psychochemical characteristics) and external (operational parameters of STPs) factors.

Relationship between wastewater parameters and antibiotic removal

The performance of the STPs in terms of wastewater parameters is shown in Table S6. In general, EAs and CAS showed higher removal efficiencies on most parameters, while IT was the least effective treatment system. The longer retention time in EAs allowed the system to minimise the sludge generation and handle variable flow rates and organic strengths

in wastewater. The efficiency of EAs has been well documented since a few decades ago (Sotirakou et al., 1999).

While IT was a popular treatment system a century ago, it is commonly used as the primary treatment nowadays. A simple anaerobic process is designed without aeration and mixing, and its typical performance is less effective than that of activated sludge systems (Reynolds and Richards 1996). Thus, it is understandable that the average removal of antibiotics by ITs across almost all parameters is lower as compared to EA plants. The removal efficiencies of most parameters for CAS systems are between those of EA and IT plants.

The negative ammonia removal in IT plants was possibly due to the result of the dissimilatory nitrate reduction to ammonia, which is a common process that happens during the denitrification process under anaerobic treatment conditions (Akunna et al. 1994). For CAS, it was unclear that the ammonia was not effectively removed. Furthermore, as organic nitrogen rapidly breaks down into ammonia in wastewater, the minimum nitrification might have been insufficiently efficient to surpass the generation of ammonia in both plants (Peavy and Tchobanoglous 1985).

Spearman's correlation (Sp) between the removal of wastewater parameters and antibiotics was computed to determine the relationship between both removals. To improve the reliability of the results, the correlation was based on the removal data from the same batch only. For example, the antibiotic removal data from October 2018 and February and June 2019 were correlated with the removal data of wastewater parameters in those respective months and batches only. The correlation analyses did not include results from the CAS plant because less than two antibiotics were detected in samples from the CAS plant, which could not generate any relationship based on the data obtained. However, the results presented in this section were statistically insignificant (ANOVA, $p > 0.05$) owing to the small sample size.

Table 3 shows the Sp values of the removals. In general, the relationship was considered strongly positively correlated if the value was above 0.7 and strongly negatively correlated if the value was below − 0.7 (RGC-IBG n.d.). Based on this criterion, only certain wastewater parameters were strongly correlated to antibiotic removal. The values that were above 0.7 and less than − 0.7 are highlighted in bold.

The removal of AMP in EA plants was strongly correlated to BOD (0.7), ammonia (0.8) and TP (− 0.7) removal. This indicated that AMP removal was higher when the removal of BOD and ammonia were higher, and inversely with TP removal in EA plants. On the other hand, the removal of AMP in IT plants was positively correlated to tCOD (0.7) and BOD (0.7). These relationships indicated that the removal of AMP was higher when the removal of tCOD and BOD were higher in IT plants. The positive relationships between AMP removal and BOD and ammonia removal in both plants showed that AMP could be cometabolised by heterotrophic and nitrifying bacteria during the treatment process. A previous study revealed similar experimental results, showing that AMP was better removed when COD removal was stronger and nitrification occurred (Labinghisa and Rollon 2014).

The removal of CIP was weakly correlated with the removal of any wastewater parameters in EA plants. However, the removal of CIP in IT plants was strongly correlated with the removal of TN (0.77) and TP (− 0.9). The positive correlation between TN and CIP removal suggested the possible cometabolism of CIP under anaerobic conditions. Although anaerobic conditions do not favour ammonia oxidation, Jetten et al. (1998) reported that ammonia could be oxidised through the anammox process. As shown in Table S6, no ammonia was oxidised during the treatment process, and since nitrate was presented in the influent of wastewater (data not shown), the denitrification process was the only process that could be associated with the removal of CIP. One of the denitrifiers (nitrate-reducing bacteria) was previously reported to be significantly related to the removal of CIP during the denitrification process (Martins et al. 2018). A study showed that CIP was removed through cometabolism during the denitrification process and consumed by nitrate-reducing bacteria as the sole carbon and electron source. Furthermore, the removal of CIP was previously reported as effective and rapid under denitrification

Table 3 Spearman correlations between removal of antibiotics and wastewater parameters

Sp values	AMP		CIP		ERY		SMX	
	EA	IT	EA	IT	EA	IT	EA	IT
tCOD	0.5	**0.7**	0.41	− 0.49	0.5	**0.73**	− 0.41	0.5
BOD	**0.7**	**0.7**	− 0.29	− 0.64	− 0.5	**0.73**	− 0.11	0.5
TSS	− 0.6	− 0.5	− 0.35	0.54	0.5	− 0.32	0.41	− 0.5
NH_3	**0.8**	− 0.2	− 0.07	0.18	**1.0**	**0.73**	− 0.07	0.5
TN	0.1	− 0.6	0.57	**0.77**	0.5	0.63	**0.77**	− 0.5
TP	**− 0.7**	− 0.4	− 0.07	**− 0.9**	0.5	**− 0.73**	− 0.63	− 0.5

conditions, indicating the occurrence of CIP cometabolism (Ooi et al. 2018).

The removal of several wastewater parameters was strongly correlated with ERY removal, including ammonia in EA plants and TCOD, BOD, ammonia and TP in IT plants. The positive correlation between ammonia and ERY removal in EA plants indicated that ERY was possibly being cometabolised by nitrifying bacteria during the treatment process of EA plants. Previous findings reported by Suárez et al. (2010) and Fernandez-Fontaina et al. (2016) revealed that ERY was effectively removed under nitrifying conditions but less effectively so without nitrifying bacteria. This proved the importance of nitrifying bacteria and their association with the removal of ERY under aerobic conditions.

The positive relationship between TCOD and BOD removal and ERY removal in IT plants indicated the importance of heterotrophic bacteria during the removal of ERY under aerobic conditions, as confirmed by Fernandez-Fontaina et al. (2016). The positive correlation between ammonia and ERY removals could be explained by the conversion of organic nitrogen into ammonia in the anaerobic process. The lower generation of ammonia could be translated as better ammonia removal. The generation of ammonia suppressed the ERY removal in IT plants, which resulted in the lower removal of ERY. Hence, the lesser the ammonia generated was, the better the removal of ERY would be.

The removal of SMX was only negatively correlated to the TN removal in EA, and no removal of the wastewater parameters was correlated with the removal of SMX in IT plants. The positive relationship between SMX removal and ammonia removal in EA plants agreed with the results highlighted by previous studies (Kang et al. 2018; Zhao et al. 2018), signifying the role of nitrifying bacteria during the treatment of SMX.

It is worth noting that the removal of most antibiotics was negatively correlated with TP removal in EA and IT plants. To the best of our knowledge, no study has been conducted concerning this observation. It is known that phosphate in wastewater is mainly degraded by phosphorus-accumulating organisms (PAO) (Dorofeev et al. 2020). Hence, it can be postulated that the removal of the antibiotics was strongly related to these microorganisms during wastewater treatment. However, further research is required to clarify this phenomenon.

Conclusions

In this work, the occurrence and removal of four types of antibiotics in three different types of STPs were studied and presented. Most selected antibiotics were detected in the influents, secondary effluents and final effluents of STPs with concentrations ranging from ng/l to µg/l levels. The STPs investigated could only partially remove the selected antibiotics from the wastewater.

In general, no evidence showed that the removal of antibiotics accomplished by conventional treatment systems was significantly superior to that of decentralised treatment systems. AMP and SMX were effectively removed by EA and CAS plants (> 75% reduction), ERY was effectively removed by IT plants (> 85% reduction) and CIP could not be removed effectively by any type of system (< 30% reduction). Therefore, the combination between conventional and decentralised treatment systems could be the possible solution for better antibiotic removal.

Using mass flow analyses, it was estimated that 7% to 147%, 2.7% to 89% and 0.3% to 18.7% of antibiotic fractions were left in the secondary effluent, final effluent and sewage sludge of EA plants, respectively. For IT plants, 21.9% to 126.3% and 0.004% to 0.3% of the antibiotic were left in the final effluent and sewage sludge, respectively. Approximately 2.2% to 106.7% of the antibiotic was left in the final effluent of the CAS plant. It was estimated that 117.38 g, 273.07 g, 170.55 g and 734.57 g of AMP, CIP, ERY and SMX, respectively, were discharged annually into the environment from the studied STPs. Mass balance analyses also showed that biodegradation was the most important removal pathway for all antibiotics in all STPs. The sorption of antibiotics only accounted for marginal antibiotic removal in all STPs.

The removal of certain wastewater parameters and antibiotics were highly correlated. Despite ANOVA analysis showing these relationships were not sufficient significant, these preliminary findings acted as the recommendation for future study.

As a conclusion, conventional and decentralised sewage treatment systems are insufficient to prevent the discharge of antibiotics into the environment. While the impacts of these chemicals on the environmental ecosystem are not well established, the increasing consumption of antibiotics that may find their way into STPs and be eventually discharged into our water bodies could pose serious threats to environmental as well as human health. As conventional and decentralised treatment systems are well adopted globally due to economic considerations, more work must be carried out to improve their performance in dealing with these emerging pollutants.

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Authors' contributions CXC performed literature search, experimental work, data analysis and wrote the original draft; AA and ELY supervised, reviewed and revised the draft; ZZN managed the project and funding.

Declarations

Conflict of interest The authors declare that they have no conflicts of interest.

Ethical approval This article does not contain any studies with human participants or animals performed by any of the authors.

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